

Vector chiral spin liquid phase in absence of geometrical frustration

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Making use of detailed classical Monte Carlo simulations, we study the critical properties of a two dimensional planar spin model on a square lattice composed by weakly interacting helimagnetic chains. We find a large temperature window where the vector chirality order parameter, $\langle \boldsymbol{\kappa}_{jk} \rangle = \langle \mathbf{S}_j \times \mathbf{S}_k \rangle$, the key quantity in multiferroic systems, takes nonzero value in absence of long-range order or quasi-long-range order, so that, our model is the first example where, at finite temperatures, a vector chiral spin liquid phase in absence of geometrical frustration is explicitly reported. We also show that the strength of interchain interaction is fundamental in order to obtain the vector chiral spin liquid phase. The relevance of our results for three-dimensional models is also discussed.

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Geometrical frustration and/or competition between interactions can lead to exotic noncollinear magnetic thermodynamic phases, which can be characterized by unusual order parameters. Particularly relevant are two order parameters: scalar chirality $\langle \chi_{jkl} \rangle = \langle \mathbf{S}_j \times \mathbf{S}_k \cdot \mathbf{S}_l \rangle$ and vector chirality (or spin current) $\langle \boldsymbol{\kappa}_{jk} \rangle = \langle \mathbf{S}_j \times \mathbf{S}_k \rangle$. These two chiralities present different symmetries: nonzero value of $\langle \chi_{jkl} \rangle$ implies that the time-reversal symmetry is broken, while parity symmetry breaking comes with $\langle \boldsymbol{\kappa}_{jk} \rangle \neq 0$. Both of them are relevant in strongly correlated electron systems: a nonzero $\langle \chi_{jkl} \rangle$ gives rise to large anomalous Hall effect [1] and leads to orbital electric currents in frustrated geometries [2], while new electromagnetic phenomena emerge in Mott insulators as a consequence of induced $\langle \chi_{jkl} \rangle$, generated by the coupling between the $\langle \boldsymbol{\kappa}_{jk} \rangle$ and an external homogeneous magnetic field [3]. On the other hand, relativistic spin-orbit interaction leads to a coupling between the vector chirality and the electric polarization [4–7] which play a fundamental role in magnetoelectric properties. This coupling permits also to obtain experimental informations about the vector chirality (which is difficult to measure owing to the absence of external physical fields that couple directly to $\boldsymbol{\kappa}_{jk}$): the chiral components in multiferroic MnWO_4 have been detected by neutron diffraction using spherical polarization analysis as a function of temperature and of external electric field [8]. The vector chirality, which is the argument of this letter, always accompanies helical magnetic order, and it can arise from spontaneous \mathbb{Z}_2 symmetry breaking in systems with competitive exchange interactions [9], or it can be stabilized by the Dzyaloshinskii-Moriya antisymmetric exchange interaction in noncentrosymmetric compounds, [6, 10, 11]. However, the vector chiral symmetry can be broken also in a magnetically disordered state. Such phase is named a vector chiral spin liquid phase and has been intensively studied in the last years. It has been predicted to occur in one-dimensional (1d) frustrated quantum magnetic systems [12–14]. For higher dimension d it is crucial to un-

derstand if the vector chiral spin liquid phase is stable also in presence of thermal fluctuations [14]. For $d=2$, this phase has been clearly obtained at finite temperature T by classical Monte Carlo (MC) simulation of a triangular lattice of spins with bilinear and biquadratic interactions [15]. However, for models without geometrical frustration and $d=2, 3$, a clear evidence of this exotic phase is yet lacking, even if Onoda and Nagaosa [16, 17], investigating a Ginzburg-Landau Hamiltonian describing helical magnets, suggest that a vector chiral spin liquid phase can be stabilized even in $d=3$. This prediction was questioned by Okubo and Kawamura [18], because the results of their classical MC simulations do not show any evidence of such phase, but only a first order phase transition to a helimagnetic order. In this context, it is important to note that in the quasi-1d XY organic magnet $\text{Gd}(\text{hfac})_3\text{NITet}$ [19] (a compound with high value of spin, $S=7/2$) a 3d vector chiral spin liquid phase has been experimentally observed. This result is due to the fact that $d=1$ is the lower critical dimension for an Ising order parameter, like $\langle \boldsymbol{\kappa}_{jk} \rangle$, so that the chiral correlation length, which diverges exponentially at low T , is much larger than the spin correlation length, which diverges with a power law. Taking into account the interchain interaction within mean field approximation a 3d vector chiral spin-liquid phase results at intermediate T [20]. However, theoretical results obtained considering also the interchain fluctuations are still lacking, and a direct numerical evidence of such a chiral phase in quasi-1d system will be relevant.

In this letter we present the results obtained by employing accurate classical MC simulation techniques to investigate a 2d spin system composed by weakly interacting helimagnetic chains. Despite the absence of geometrical frustration (the model being defined on a square lattice), a clear separation can be observed between the chiral phase transition temperature T_κ and the Kosterlitz-Thouless (KT) one T_{KT} separating the quasi-long-range spin ordered phase from the disordered one.

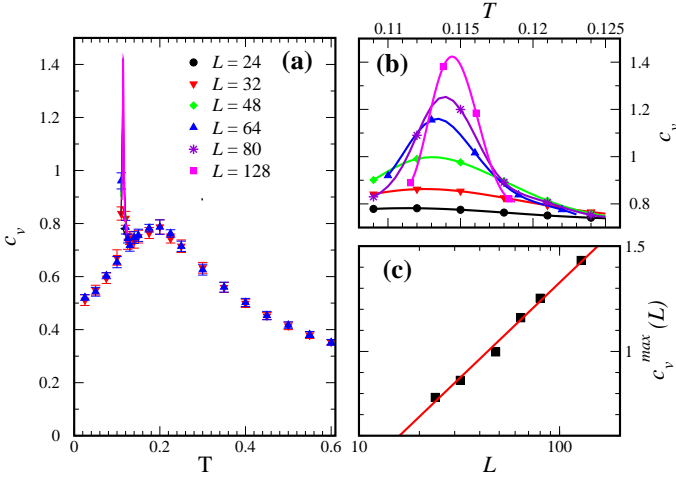


FIG. 1: (Color online) (a) Specific heat *vs.* T for the model (1) with $J'=0.1J$ for different L (see legend). (b) MH result around T_κ for different L . (c) Maximum of the specific heat *vs.* L (error bars lie within point size). Continuum line: fit function, see text.

We consider a simple square lattice on the (x, y) -plane composed of $N=L \times L$ planar spins $\vec{S}_{i,j}$ ($|\vec{S}|=1$), whose interactions are described by the Hamiltonian:

$$\mathcal{H} = - \sum_{i=1}^L \sum_{j=1}^L \left\{ J_1 \vec{S}_{i,j} \cdot \vec{S}_{i,j+1} + J_2 \vec{S}_{i,j} \cdot \vec{S}_{i,j+2} + J' \vec{S}_{i,j} \cdot \vec{S}_{i+1,j} \right\} ; \quad (1)$$

j labels spins along each chain, while i is the chain label. Intra-chain exchange interactions are ruled by a nearest neighbour (NN), ferromagnetic (FM) coupling constant J_1 and a next-nearest neighbour (NNN), antiferromagnetic (AFM) coupling J_2 ; interchain NN spin interactions are ruled by the FM coupling constant J' . If the condition $\delta \equiv |J_2|/J_1 > 1/4$ is fulfilled, the ground state corresponds to a helical order along the chains, with a pitch vector $q_{\parallel} = \pm \cos^{-1}(-1/4\delta)$. In the following we take $\delta = 0.3$ (i.e. $J_1=1$, and $|J_2|=0.3$), while T will be given in units J_1 . Periodic boundary conditions have been applied along the direction perpendicular to the chains while, due to the incommensurate helix modulation, free boundary conditions were taken along the chain direction. Configuration sampling has been carried on making use of the usual Metropolis technique, while correlations between sampled configurations were mitigated by microcanonical over-relaxed moves[21]. For each T at least three different runs have been performed, each run being composed of 24×10^6 MC sweeps, with the first 4×10^6 thermalization steps being discarded. Near the critical regions the multiple-histogram (MH) methods [21] were employed. Results for $L = 24 - 128$ are reported.

The weakly interacting chain system has been investi-

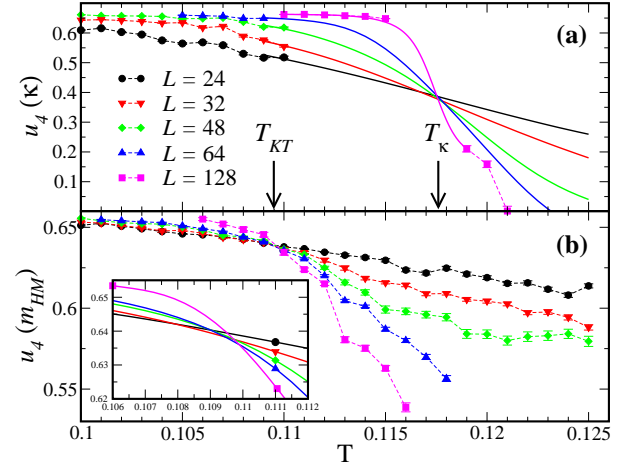


FIG. 2: (Color online) (a) Binder cumulant for the order parameter κ *vs.* T for different lattice sizes; continuous lines: MH interpolation. (b) Binder cumulant for the helical order parameter *vs.* temperature for different lattice sizes. Inset: MH interpolation around T_{KT} .

gated by setting $J'=0.1J$: In Fig. 1a the obtained results for the specific heat, c_v , *vs.* T are reported. We observe a well defined narrow and sharp peak at $T \simeq 0.12$, which should be ascribed to the onset of a vector chiral spin liquid phase. In this T range, the scaling behavior of c_v for different L is reported in Fig. 1b: We immediately observe as increasing L the peak more and more acquires the typical features associated with a second order phase transition in the thermodynamic limit. Increasing T , a second broad and size-independent peak is observed at $T \simeq 0.2$, which is consistent with a KT scenario. In order to estimate the critical temperatures, we employ the Binder's fourth cumulant $u_4 = 1 - \langle \mathcal{O}^4 \rangle / 3 \langle \mathcal{O}^2 \rangle^2$ [21], where \mathcal{O} will be the chirality, $\kappa = K' \sum_{ij} [\vec{S}_{ij} \times \vec{S}_{ij+1}]^z$ (where $K' = [L(L-1) \sin q_{\parallel}]^{-1}$), or the helical order parameter, m_{HM} , defined as $m_{HM} = K'' \int dq_{\parallel} S(\vec{q})$, where $S(\vec{q})$ is the structure factor, with $\vec{q} = (0, q_{\parallel})$, and the normalization factor K'' is the reciprocal of the structure factor integral at $T=0$ [22]. The Binder cumulant for different L is reported in Fig. 2a and b for the chirality and the helical order parameters, respectively. For the chirality we can evaluate $T_\kappa = 0.1176(6)$, while for the helical order parameter, we obtain $T_{KT} = 0.1095(5)$. The data in Fig. 2 allows us to assert that the two critical temperatures are well distinguishable with $(T_\kappa - T_{KT})/T_{KT} \simeq 7.4\%$. The identification of the crossing temperature in Fig. 2b as the KT transition temperature can be further validated by making use of the finite-size scaling (FSS) relation $\chi_m(L) \propto L^{\gamma/\nu}$; a best fit procedure gives $\gamma/\nu = 1.77(3)$ (Fig. 3a), fully consistent with the KT behaviour of a $2d$ planar system. Another evidence of the presence of two distinct critical points comes from the scrutiny of

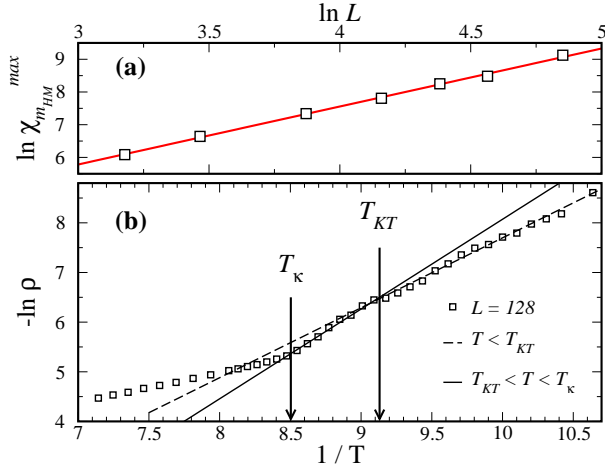


FIG. 3: (Color online) (a) Logarithm of the maximum of $\chi_{m_{HM}}$ as a function of $\ln L$. The error bars lie within the symbols. (b) Vortex density ρ vs. T^{-1} for $L=128$, error bars lie within point size. Continuum line: linear regression in the chiral region. Dashed line: linear regression in the KT regime.

the vortex density, ρ [28]. In the dilute-gas approximation, we have $\rho \sim \exp(-2\mu/T)$, where 2μ is the energy required to create a pair of vortices [23], and it can be obtained by linear fit of $-\ln \rho$ as a function of T^{-1} (Fig. 3b). Three different regimes can be identified: low- T ($T < T_{KT}$), intermediate- T ($T_{KT} < T < T_\kappa$), and high- T regime ($T_\kappa < T$). The linear fit in the range $T_{KT} < T < T_\kappa$ (solid line) shows an activation energy of dissociated vortex pairs greater than that obtained for lower temperatures $T < T_{KT}$, where all vortex pairs are bounded, with a clear slope change at $T \simeq T_{KT}$. Finally, the creation of other dissociated vortex pairs appears again easier in the region $T > T_\kappa$, where μ strongly decreases signaling the onset of a complete disorder [24].

A proper characterization of the vector chiral spin liquid transition involves several aspects. A first issue, concerning the order of the transition, can be coped with by analyzing the equilibrium energy distribution at $T=T_\kappa$. Even at the largest simulated size of the lattice, $L=128$, no double-peaked structure was observed, so that we have no explicit indication in favor of classifying the chiral transition as a first-order one.

The universality class pertaining to the vector chiral spin liquid transition has been investigated by an accurate FSS analysis. In Fig. 4a the chiral susceptibility, χ_κ , is displayed for different values of L . From the expected dependence of the peak position temperature on L , $T_\kappa(L) = T_\kappa + cL^{-1/\nu}$, we can estimate the critical exponent ν , making use of the value of T_κ previously obtained from the Binder's cumulant discussed above, getting $\nu=1.02(5)$ (Fig. 4b). Analyzing the peak values of χ_κ with the FSS relation $\chi_\kappa(L) \propto L^{\gamma/\nu}$, we obtain the ratio

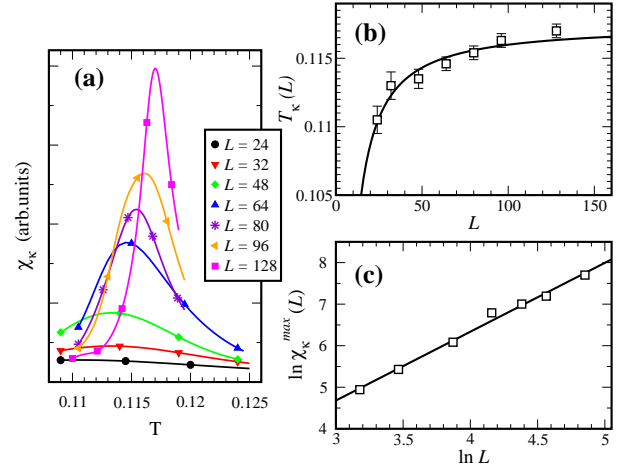


FIG. 4: (Color online) (a) Chiral susceptibility vs. T by MH interpolation for different sizes. In (b) the temperature where the peak of χ_κ is located is reported vs. L . In (c) the maximum value of χ_κ is reported as a function of $\ln L$. Error bars fall within the symbols.

$\gamma/\nu=1.66(7)$ (Fig. 4c), which implies $\gamma=1.70(8)$. These values of γ and ν are in very fair agreement with $\gamma=7/4$ and $\nu=1$, i.e. the proper values of the Ising universality class in $2d$. Concerning the critical exponent α for c_v , which for the Ising universality class in $2d$ is 0, the c_v -peak values, $c_v^{max}(L)$, vs. L are very well fitted (see Fig. 1c) by the FSS relation proper of the $2d$ Ising model $c_v^{max}(L) = A + B \ln(L) + CL^{-1}$, [25]. This, allows us to conclude that $\alpha=0$, confirming the Ising character of the vector chiral spin liquid transition.

We point out that the quasi-1d nature of the model is fundamental in order to obtain a vector chiral spin liquid phase in absence of (quasi-)long-range order. This has been explicitly checked by MC simulations we have performed assuming $J'=J_1$, a model investigated by Garel and Doniach [26] many years ago. In Fig. 5a-d a summary of the obtained results is reported.

For the specific heat, c_v , reported vs. T , we observe a size-independent broad peak at $T \simeq 0.75$, consistent with a KT scenario, while a size-dependent, narrow peak at $T \simeq 0.34$ is found. Using the helicity modulus $\Upsilon(T)$ [27], one is able to evaluate T_{KT} taking advantage of the universal jump: $\Upsilon(T)/T \rightarrow 2/\pi$ for $T \rightarrow T_{KT}$. We estimate $T_{KT} \simeq 0.45$. For the largest simulated size ($L=108$), χ_κ shows a narrow peak at the same temperature T_κ of the narrow, size-dependent, c_v peak and, above all, T_κ is significantly lower than T_{KT} . These observations are corroborated by the data obtained for a new parameter defined as $M = \frac{1}{L} \sum_{i=1}^L m_i$ where $m_i = \sqrt{(\frac{1}{L} \sum_{j=1}^L S_{i,j}^x)^2 + (\frac{1}{L} \sum_{j=1}^L S_{i,j}^y)^2}$ is the columnar magnetization perpendicular to the helical displacement. This observable turns out to be relevant both for the

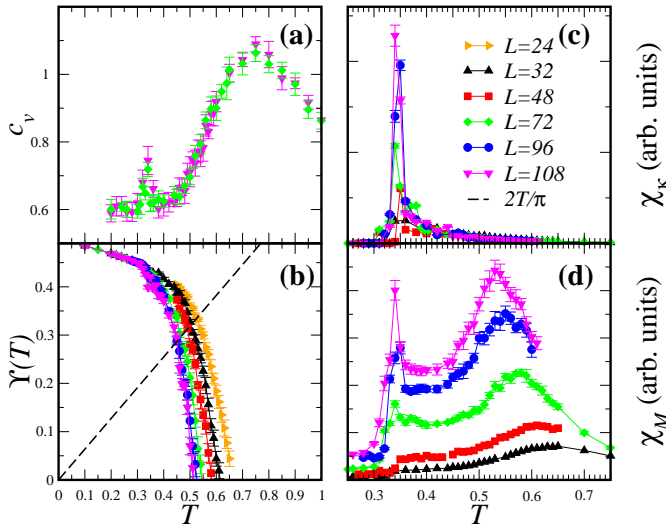


FIG. 5: (Color online) Observables calculated for the system with $J_1=J'$ for different size L : (a) specific heat; (b) helicity modulus; (c) chiral susceptibility; (d) susceptibility of M (see text).

chiral phase and for the establishment of the KT phase: Indeed, both two- and four-points correlations contribute to its susceptibility χ_M (Fig. 5d), which displays a first anomaly, which progressively stabilizes at T_{KT} when L increases, signaling the onset of a quasi-order; subsequently, at lower- T , χ_M has a second anomaly consistent with those already displayed by c_v and χ_k . So, we can definitely estimate $T_\kappa \simeq 0.34$. We conclude that, for $J'=J_1$, we have a clear separation between the KT behaviour and chiral setup, but, at variance with the quasi-1d case, the onset of the chiral order is established at a temperature T_κ lower than T_{KT} .

In conclusion, we have presented the outcomes of intensive MC simulations for a 2d XY classical spin system, defined on a square lattice, composed by weakly interacting frustrated chains. We observe a clear separation between the vector chiral spin liquid phase and the quasi-long-range ordered phase, with $T_\kappa > T_{KT}$. We have found that, in a system without geometrical frustration, the chirality displays a second order phase transition, consistent with the 2d Ising universality class. This result confirms the intriguing possibility of an emergent finite-temperature phase showing chiral long range order in the absence of the helical one as investigated by many authors in the multiferroic context [14–17]. We found the quasi-1d nature of the system being fundamental in order to observe such an exotic phase in absence of (quasi)-long-range order: indeed, assuming the same NN exchange constants in both directions ($J'=J_1$) we find that the sequence of phase transitions can be reversed $T_\kappa < T_{KT}$. The opposite sequence of the two phase transitions for the investigated 2d models can also give

indications for the behavior of their 3d counterparts. As we move from $d=2$ to $d=3$, the KT phase transition is replaced by a proper second order phase transition to a helimagnetic spin arrangement which implies an underlying chiral one, so that it is not surprising that in their MC simulation Okubo and Kawamura [18] do not observe a chiral phase but only the phase transition to the helical order, which also entails chiral order. On the contrary, for a 3d collection of weakly interacting helimagnetic chains, it is reasonable to hypothesize that the phase transition to helimagnetic order occurs at a lower T than the chiral one, and the vector chiral spin-liquid phase can manifest [20] according to the scenario recently emerged from experiments on the quasi-1d organic high-spin magnet $\text{Gd}(\text{hfac})_3\text{NITet}$ [19].

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